Efficient cross-polarized wave generation with holographic cut crystals for femtosecond laser contrast filtering

S. Kourtev[a,*], L. Canova[b], N. Minkovski[a], A. Jullien[b], O. Albert[b], R. Lopez-Martens[b], and S. M. Saltiel[a]

[a] Faculty of Physics, Sofia University, 5 J. Bourchier Blvd., Sofia-1164, Bulgaria;

[b] Laboratoire d'Optique Appliquée, Ecole Nationale Supérieure de Techniques Avancées, Ecole Polytechnique, CNRS-UMR 7639, 91761 Palaiseau Cedex, France

ABSTRACT

We show both theoretically and experimentally that cross-polarized wave generation (XPWG) is more efficient when the input fundamental beam propagates along the [011] direction in a cubic crystal than along the previously used [001] direction. With a [011]-cut BaF_2 crystal we measured the highest to date XPWG conversion efficiencies. We prove another very important advantage of the [011]-cut approach: weak induced phase mismatch and consequently no need for its compensation.

Keywords: cross-polarized wave generation, contrast improvement of femtosecond lasers

1. INTRODUCTION

Cross-polarized wave generation (XPWG) is a four-wave mixing process governed by the anisotropy of the real part of the crystal's third order nonlinear tensor $\chi^{(3)}$. It has attracted an interest in recent years primarily as a method to increase the temporal contrast of femtosecond pulses,[1–3] which is crucially important for the interaction of ultra-intense femtosecond laser radiation with solid-state targets. XPWG has proved to be an effective and robust contrast-enhancement technique that allowed for increasing of the contrast ratio of energetic femtosecond laser pulses up to 10–11 orders of magnitude.[1–3]

The XPWG is an automatically phase-matched process, but at high intensities the exact phase matching is broken since self- and cross-phase modulation introduce additional nonlinear phase shift, thus leading to saturation of the XPWG. This induced phase mismatch usually is compensated by changing the orientation of the input polarization. All efficient schemes realized so far are with [001]-cut (z-cut) BaF_2 samples. It is of great interest to search for new types of crystals and/or for different orientations that can yield higher efficiencies. Here we report for the first time the experiment with [011]-cut (holographic cut) BaF_2 samples that proved to be 30% more efficient cross-polarized wave generator than the previously used z-cut thus opening the way to better usability of XPWG contrast filtering in chirped-pulse amplification (CPA) femtosecond lasers. Furthermore, the induced intensity-dependent phase mismatch is negligible for [011]-cut crystals in contrast to the strong dependence on the orientation of the input polarization observed with z-cut samples. Therefore in [011]-cut configuration there is no need for crystal (or equivalently the input polarization) rotation when changing the input intensity.

BaF_2 is a cubic crystal belonging to m3m symmetry point-group. Crystals belonging to other symmetry groups have also been investigated. In Ref. 4 an YVO_4 crystal is used as XPW generator pumped along its optical axis (z-axis). Although the nonlinearity of YVO_4 is higher, the maximum XPW efficiency that can be achieved is the same. Moreover, because of its linear birefringence, (i) a very accurate crystal alignment is mandatory in order to avoid degradation of the input polarization, and (ii) z-cut is the only permitted crystal orientation for the same reason. There are no reports about other crystals suitable for XPWG. Another solution is to explore more efficient orientations in cubic crystals.

*Electronic mail: skourtev@phys.uni-sofia.bg

15th International School on Quantum Electronics: Laser Physics and Applications, edited by Tanja Dreischuh
Elena Taskova, Ekaterina Borisova, Alexander Serafetinides, Proceedings of SPIE Vol. 7027, 70271Q
 · doi: 10.1117/12.822540

2. THEORETICAL PART

For cubic crystals (m3m) the efficiency of the XPWG depends on the product of the $\chi^{(3)}_{xxxx}$ component, the crystal length L, the input intensity, and the anisotropy of $\chi^{(3)}$-tensor $\sigma = \left(\chi^{(3)}_{xxxx} - 3\chi^{(3)}_{xxyy}\right)/\chi^{(3)}_{xxxx}$.[4–7] If we denote with A the complex amplitude of the fundamental beam and with B the complex amplitude of the cross-polarized wave (XPW) then the following system of ordinary differential equations may be derived in plane-wave approximation:

$$\frac{dA(\zeta)}{d\zeta} = i\gamma_1 AAA^* + i\gamma_2 AAB^* + 2i\gamma_2 ABA^* + 2i\gamma_3 ABB^* + i\gamma_3 BBA^* + i\gamma_4 BBB^*, \tag{1}$$

$$\frac{dB(\zeta)}{d\zeta} = i\gamma_5 BBB^* + i\gamma_4 BBA^* + 2i\gamma_4 ABB^* + 2i\gamma_3 ABA^* + i\gamma_3 AAB^* + i\gamma_2 AAA^*, \tag{2}$$

where ζ is the longitudinal coordinate in the direction of light propagation, and the asterisk superscript, as usual, means complex conjugate quantity. Nonlinear coupling coefficients γ_1 through γ_5 in Eqs. (1) and (2) depend in a complex manner on the crystal orientation (i.e. on the direction of light propagation in the crystal) and on the polarization of the input light. Theoretical analysis of the XPWG efficiency for arbitrary orientation of the input fundamental beam shows that for cubic crystals the holographic cut ensures the highest efficiency since $|\gamma_2|$ and $|\gamma_4|$ reach their maximal values when the input beam propagates along [011], [101], or [110] direction in the crystal. γ-coefficients for both the commonly used z-cut and the proposed most efficient holographic cut are summarized in Table 1. γ_1 and γ_5 are responsible for the self-phase modulation (SPM) of the two waves A

Table 1. Nonlinear coupling coefficients used in Eqs. (1) and (2) for z-cut and for the holographic cut. $\gamma_0 = 6\pi\chi^{(3)}_{xxxx}/8n\lambda$.

	z-cut	holographic cut
γ_1	$\gamma_0\left[1-(\sigma/2)\sin^2(2\beta)\right]$	$\gamma_0\left[1-(\sigma/4)\cos(2\beta)+(3\sigma/16)\cos(4\beta)-7\sigma/16\right]$
γ_2	$-\gamma_0(\sigma/4)\sin(4\beta)$	$\gamma_0\left[(\sigma/8)\sin(2\beta)-(3\sigma/16)\sin(4\beta)\right]$
γ_3	$\gamma_0\left[1/3-(\sigma/4)\cos(4\beta)-\sigma/12\right]$	$\gamma_0\left[1/3-(3\sigma/16)\cos(4\beta)-7\sigma/48\right]$
γ_4	$\gamma_0(\sigma/4)\sin(4\beta)$	$\gamma_0\left[(\sigma/8)\sin(2\beta)+(3\sigma/16)\sin(4\beta)\right]$
γ_5	$\gamma_0\left[1-(\sigma/2)\sin^2(2\beta)\right]$	$\gamma_0\left[1+(\sigma/4)\cos(2\beta)+(3\sigma/16)\cos(4\beta)-7\sigma/16\right]$

and B, respectively. γ_2 and γ_4 govern the process of XPWG from A to B through the last term of Eq. (2) and from B to A through the last term of Eq. (1). Note that SPM and XPWG are generally different for the two waves A and B except for the special case of z-cut, where, because of the rotational (around z-axis) symmetry, the following relations hold true: $\gamma_5 = \gamma_1$ and $\gamma_4 = -\gamma_2$.

The dependence of the real part of the nonlinear coupling coefficient that determines XPWG (γ_2 in our case) on the polarization of the fundamental beam is shown in Fig. 1(a) for both the commonly used z-cut and the most efficient holographic cut. For both cuts β is the angle between the polarization direction of the input fundamental beam and the crystalline axis x. In calculations $\gamma_0 = 1$ and $\sigma = -1.2$ (a typical for the anisotropy of BaF_2 value[4,8,9]) were used. In non-depleted regime the efficiency of XPWG is proportional to the squared XPW nonlinear coupling coefficient.[4] As seen from Fig. 1(a) the maximum absolute value of γ_2 for [011]-cut is 12.22% greater than the maximum for z-cut and hence we may expect almost 26% increase in the XPWG efficiency. This advantage does not depend on the particular choice of the values of γ_0 and σ since γ_2 depends linearly on them for arbitrary crystal orientation (i.e. $\gamma_2 \propto \gamma_0\sigma$; see also Table 1).

We numerically solve the system of differential equations (1) and (2) in order to find the XPWG efficiency. In the case of plane-wave propagation the efficiency is defined as the ratio of the intensities of the XPW and the input fundamental wave. Calculated XPWG efficiencies for the new [011]-cut (holographic cut) and for the [001]-cut (z-cut) are shown in Fig. 1(b). The dimensionless argument $S = \gamma_0 |A_0|^2 L$, where $A_0 = A(\zeta = 0)$ and L is the crystal length, is proportional to the product nonlinearity × input intensity × crystal length. The initial condition for the B-wave is $B(0) = 0$. β-angles used in calculations correspond to the angles at which $|\gamma_2|$ is

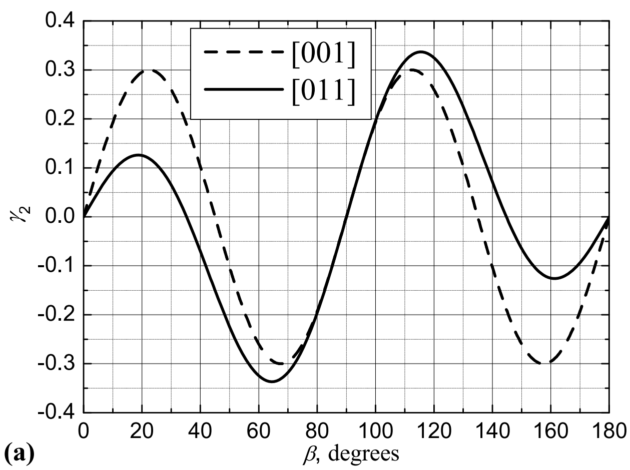

Figure 1. **(a)** Dependence of γ_2 on orientation of input polarization for the two cuts indicated. $\gamma_0 = 1$ and $\sigma = -1.2$. **(b)** Plane-wave XPWG efficiency. In calculations $\beta = 22.5°$ for [001]-cut and $\beta = 115.5°$ for [011]-cut were used. These angles correspond to the maxima of $|\gamma_2|$. See text for the definition of S.

maximum for each cut [see Fig. 1(a)]. These angles are optimal for XPW generation at low input intensity. It is, however, well known that optimum β depends on the input intensity.[4] The necessity of tuning β with the change in input intensity increases the experimental complexity in applications where, in order to avoid any parasitic SPM effects in air, the nonlinear crystal must be placed under vacuum. Below we will show that β_{opt} dependence is very weak for holographic-cut crystals [see Fig. 3(b)]. The insensitivity of β_{opt} on the input intensity for [011]-cut scheme is explained by the fact that compared to the z-cut case the phase matching conditions for optimal phase shift between the two waves ($\pi/2$) are maintained over a bigger range of input intensity. This insensitivity also means less distortions of the temporal and spatial shapes of the generated cross-polarized beam.

3. EXPERIMENTS

The experimental arrangement is shown in Fig. 2. It consists of one or two BaF_2 samples sandwiched between two uncoated crossed Glan polarizers. Both the BaF_2 samples and the polarizers were uncoated. Linearly polarized

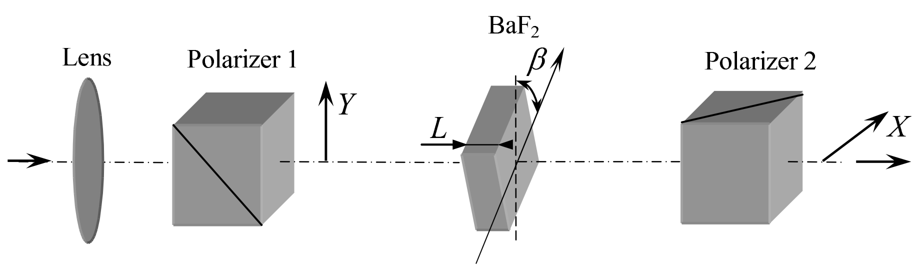

Figure 2. Experimental setup.

input fundamental beam is focused with a lens into the BaF_2 sample. In Fig. 2 Y depicts the polarization direction of the input beam and X – the polarization direction of the orthogonally polarized (XPW) output beam.

The β-dependence measurements were performed using the second harmonic of a colliding-pulse mode-locked (CPM) dye laser (620 nm, 100 fs, 10 Hz). Experimental β dependences of XPWG for z-cut and for holographic-cut samples are shown in Fig. 3(a) and Fig. 3(b), respectively. Pulses with an energy of up to 20 μJ were focused into the nonlinear crystal with a f = 150 mm lens for the z-cut sample and with a f = 500 mm lens for the holographic cut sample. Continuous curves in Fig. 3 are theoretical curves. They were obtained by numerically

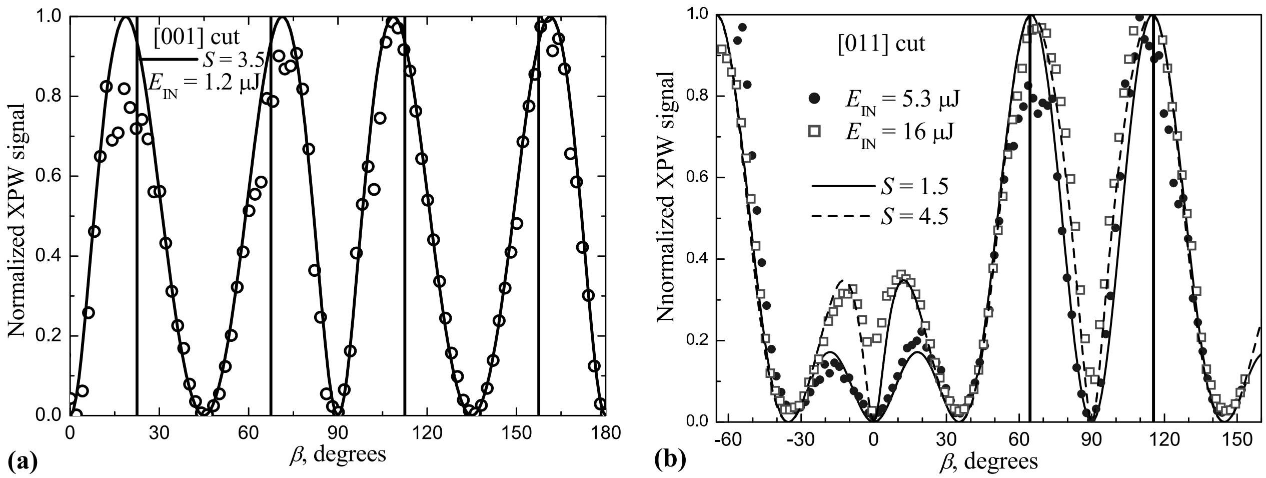

Figure 3. Normalized XPW signal as function of angle β: **(a)** for z-cut, and **(b)** for the holographic cut for two different input energies. See text for details.

solving the system of Eqs. (1) and (2) assuming Gaussian spatial and temporal profiles of the fundamental radiation. The agreement with the experiments is very good. As predicted by the theory, the two β positions (within rotation range of 180°) that correspond to maximum efficiency of the XPWG for the holographic cut are insensitive to the changes in the input energy. Vertical solid lines in Fig. 3 mark the optimal β positions for low input energy ($S < 1$). As seen from Fig. 3 optimal β does not shift with holographic cut crystals for the whole reasonable change of S while for z-cut the shift exceeds 4° even at $S = 3.5$.

The measurements of the XPWG efficiency were performed using a commercial femtosecond laser (Femtolasers GmbH) delivering up to 1 mJ, 30 fs pulses at 1 KHz and $\lambda = 800$ nm. The spatial shape of the input fundamental beam is very close to Gaussian, as shown in the inset in Fig. 4. The pulse-to-pulse stability of the laser used in the experiment is below 1% thus ensuring a very small error (3%) in the efficiency measurements. The measured conversion efficiencies (without corrections for losses) for the experiments with single 2-mm thick BaF_2 crystals are shown in Fig. 4. The input femtosecond pulses were focused with a $f = 50$ cm lens into the nonlinear crystal. The efficiency with [011]-cut BaF_2 sample saturates at 11.4%. This value can be compared with the maximum efficiency of 10% reported before with a single crystal.[4,10] Accounting for losses of the output Glan and the nonlinear crystal, we calculated a 15% internal efficiency. It is very important to note that the efficiency curve for the [001]-cut (circles) in Fig. 4 was obtained with optimization of angle β for each input energy, while for the experiment with the [011]-cut (squares) such optimization was not necessary as predicted by the theoretical model.

As the double-crystal XPWG filtering is very useful at higher energies in double CPA systems, we also tested a double-crystal scheme[2,11] for XPWG using two holographic cut BaF_2 crystals. The fundamental beam with 7 mm diameter was focused with a $f = 5$ m lens. The two crystals (placed in vacuum chamber) were set ≈ 50 cm apart, in accordance with the experimental dependence pointed out in Ref. 12. Obtained XPW energies and efficiencies are shown in Fig. 5.

The output spectrum has a nearly Gaussian shape with 80 nm full width at half maximum (for an input width of 40 nm). The maximum obtained efficiency of 29% for a 190 μJ input pulse energy yields a 55 μJ XPW pulse energy. This is the highest XPW energy efficiency recorded so far with this kind of setup. This value is in fact 1.3 times higher than the previously published[3,11,13] values obtained with z-cut crystals, which is in accordance with the prediction of the model. This result corresponds to a 50% intensity conversion from one polarization to the other. Highest efficiencies are obtained for input energies just below the continuum generation threshold.

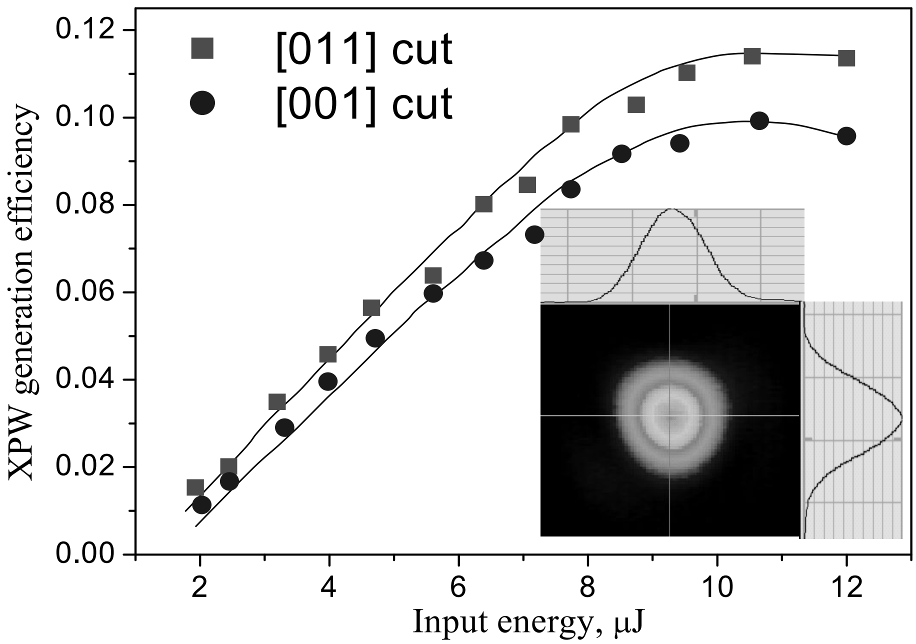

Figure 4. Comparison of measured XPWG efficiencies with single nonlinear crystal scheme with BaF_2 sample for the two cuts indicated. XPWG efficiencies are not corrected for the losses from uncoated surfaces. Continuous curves are guides for the eye. Inset: spatial shape of the input fundamental beam.

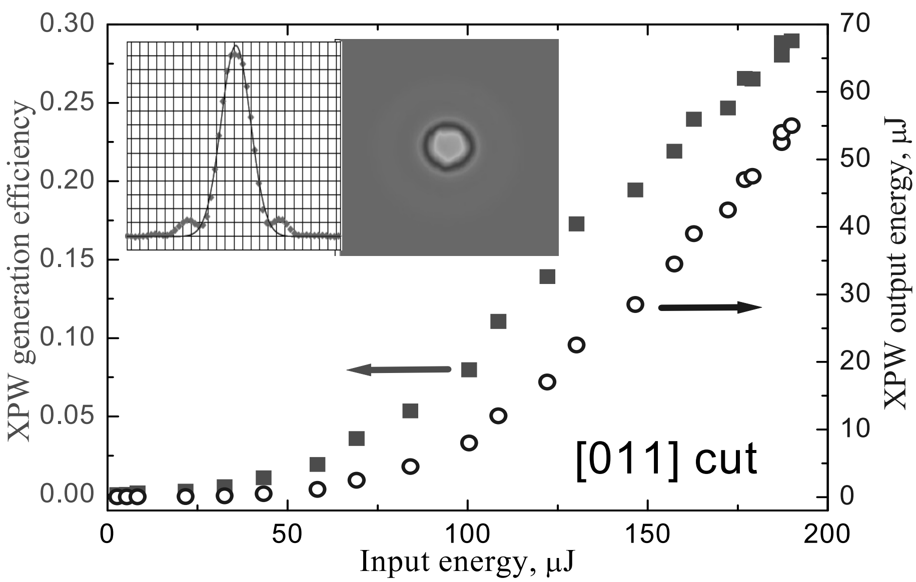

Figure 5. XPWG energy (circles) and efficiency (squares) for two-crystal setup. Inset: output XPW spatial beam shape.

4. CONCLUSION

In conclusion, we demonstrate record efficiencies for XPWG using holographic [011]-cut BaF_2 crystals. We also demonstrate that when [011]-cut crystals are used for XPWG, intensity dependent β compensation of the phase mismatch is not required. This feature makes the holographic cut easier to use for contrast filters in double CPA schemes where the nonlinear crystal must be placed under vacuum. We believe that the use of the holographic-cut cubic crystals for XPWG will stimulate the construction of more efficient and more reliable devices for temporal contrast improvement of femtosecond pulses.

ACKNOWLEDGMENTS

The support by "LaserLab-Europe" through contract *RII3-CT-2003-506350* and by the Bulgarian National Science Foundation through grant *IRNI-17* is gratefully acknowledged. S. K., N. M., and S. S. thank the Laboratoire d'Optique Appliquée for their hospitality and support.

REFERENCES

[1] Jullien, A., Albert, O., Burgy, F., Hamoniaux, G., Rousseau, J.-P., Chambaret, J.-P., Augé-Rochereau, F., Chériaux, G., Etchepare, J., Minkovski, N., and Saltiel, S. M., "10^{-10} temporal contrast for femtosecond ultraintense lasers by cross-polarized wave generation," Opt. Lett. 30, 920–922 (2005).

[2] Jullien, A., Albert, O., Chériaux, G., Etchepare, J., Kourtev, S., Minkovski, N., and Saltiel, S. M., "Highly efficient temporal cleaner for femtosecond pulses based on cross-polarized wave generation in a dual crystal scheme," Appl. Phys. B 84, 409–414 (2006).

[3] Chvykov, V., Rousseau, P., Reed, S., Kalinchenko, G., and Yanovsky, V., "Generation of 10^{11} contrast 50 TW laser pulses," Opt. Lett. 31, 1456–1458 (2006).

[4] Minkovski, N., Petrov, G. I., Saltiel, S. M., Albert, O., and Etchepare, J., "Nonlinear polarization rotation and orthogonal polarization generation experienced in a single-beam configuration," J. Opt. Soc. Am. B 21, 1659–1664 (2004).

[5] Svirko, Y. P. and Zheludev, N. I., [Polarization of Light in Nonlinear Optics], Wiley, New York (1998).

[6] Minkovski, N., Saltiel, S. M., Petrov, G. I., Albert, O., and Etchepare, J., "Polarization rotation induced by cascaded third-order processes," Opt. Lett. 27, 2025–2027 (2002).

[7] Dabbicco, M., Fox, A. M., von Plessen, G., and Ryan, J. F., "Role of $\chi^{(3)}$ anisotropy in the generation of squeezed light in semiconductors," Phys. Rev. B 53, 4479–4487 (1996).

[8] Sheik-Bahae, M., Said, A. A., Wei, T. H., Hagan, D. J., and Van Stryland, E. W., "Sensitive measurement of optical nonlinearities using a single beam," IEEE J. Quantum Electron. 26, 760–769 (1990).

[9] DeSalvo, R., Sheik-Bahae, M., Said, A. A., Hagan, D. J., and Van Stryland, E. W., "Z-scan measurements of the anisotropy of nonlinear refraction and absorption in crystals," Opt. Lett. 18, 194–196 (1993).

[10] Gordienko, V. M., Mikheev, P. M., and Syrtsov, V. S., "Nonlinear rotation of the polarization of the intense femtosecond laser radiation in BaF_2," Bull. Russ. Acad. Sci.: Phys. 71, 122–125 (2007).

[11] Jullien, A., Albert, O., Chériaux, G., Etchepare, J., Kourtev, S., Minkovski, N., and Saltiel, S. M., "Two crystal arrangement to fight efficiency saturation in cross-polarized wave generation," Opt. Express 14, 2760–2769 (2006).

[12] Albert, O., Jullien, A., Etchepare, J., Kourtev, S., Minkovski, N., and Saltiel, S. M., "Comment on 'Generation of 10^{11} contrast 50 TW laser pulses'," Opt. Lett. 31, 2990–2992 (2006).

[13] Cotel, A., Jullien, A., Forget, N., Albert, O., Chériaux, G., and Le Blanc, C., "Nonlinear temporal pulse cleaning of a 1-μm optical parametric chirped-pulse amplification system," Appl. Phys. B 83, 7–10 (2006).